N-63-4-3

RYPTON FILLED THERMIONIC CONVERTER

QUARTERLY TECHNICAL PROGRESS REPORT NO. 2
JULY 1963
AF 33(657)-10131

Aeronautical Systems Division
Air Force Systems Command
United States Air Force
Wright-Patterson Air Force Base, Ohio

ADVANCED DEVELOPMENTS DEPARTMENT
UNION CARBIDE CORPORATION
PARMA RESEARCH CENTER
PARMA, OHIO

KRYPTON FILLED THERMIONIC CONVERTER

PARMA RESEARCH LABORATORY
ADVANCED DEVELOPMENTS DEPARTMENT
UNION CARBIDE CORPORATION

QUARTERLY TECHNICAL PROGRESS REPORT NO. 2

April 1, 1963 - June 30, 1963

CONTRACT NO. AF 33(657)-10131
Project No. 8173
Task No. 817305

July 1963

AERONAUTICAL SYSTEMS DIVISION
AIR FORCE SYSTEMS COMMAND
UNITED STATES AIR FORCE
WRIGHT-PATTERSON AIR FORCE BASE, OHIO

FOREWORD

This report was initiated by the Air Force Aero Propulsion Laboratory Flight Vehicle Power Division (ASRPP-20), Air Force Systems Command. The research work on which this report is based was accomplished by Parma Research Laboratory, 'dvanced Developments Department, Union Carbide Corporation, P. O. Box 6116, Cleveland 1, Ohio, under Air Force Contract AF 33(657)-10131 entitled "Krypton Filled Thermionic Converter."

The report was written by R. Forman, Project Leader. The following scientists, engineers, and technicians have contributed to this project: J. A. Ghormley, Group Leader; F. W. Meszaros, J. R. Reiss and J. A. Raley.

Mr. P. J. Hutchison, Flight Vehicle Power Division (ASRPP-20, Ext. 22208), is project engineer for the Air Force on this program.

TABLE OF CONTENTS

ABSTRACT

Research work on irradiated inert gas thermionic diodes for the second quarter of 1963, under Contract AF 33(657)-10131, [Task 817305-26] is presented. Tubes containing both argon and krypton gases have been tested in the radiation field of a 5 megawatt swimming pool type reactor. The pressure of the gas was varied in order to determine optimum conditions for operation. The results of these measurements are described and evaluated. Differences have also been observed on irradiated diodes having the same gas and pressure but different cathode-anode spacings. The differences, in contradistinction to cesium diodes, show that small cathode-anode spacing is not particularly advantageous. A proof in principle experiment is also described showing how an inert gas filled irradiated thermionic diode can be used as an a.c. thermionic generator.

1.0. INTRODUCTION

For the last three years the Union Carbide Corporation has been engaged in a research program to study electron propagation through high pressure inert gas filled diodes. These measurements have been made with the gas both in the ionized and un-ionized state. The results of these studies have shown: (1) an inert gas filled thermionic converter, in which the space charge is neutralized by positive ions produced in the gas by nuclear radiation, seems feasible, and (2) inert gas filled thermionic diodes or converters can be designed to have negative resistance characteristics which could be used for obtaining a.c. outputs.

Government sponsorship of this continuing program started in January, 1963. The two principal objectives of this program are: (1) to design experiments to increase power output and decrease plasma resistance between cathode and anode in an irradiated inert gas diode, and (2) studies on irradiated inert gas diodes having negative resistance.

Measurements to attain the former objective are being made on thermionic diodes operating in a nuclear reactor environment, at a cathode temperature of approximately 1100°C. These tubes are cylindrical diodes employing a Philips' type cathode, which is indirectly heated by a tungsten filament, and a molybdenum anode. The standardization on this diode design permits one to evaluate the effectiveness other parameters have on the current output of the diode. The variable parameters used or to be used in these studies are:

(a) nuclear radiation dosage or intensity
(b) cathode-anode spacing
(c) type of inert gas or gases
(d) pressure of gas

During the first quarter of this contract, a number of these parameter variations were studied. Measurements of current vs. voltage output for argon filled tubes have shown that at radiation dosages of 10^8 - 10^9 rads/hr cathode current densities of about 0.2 ampere/cm^2 have been obtained. It has also been shown that the current at a given applied anode voltage shows a continued increase with increasing radiation up to the highest radiation dosage. Studies were also made on the variation of diode output with cathode-anode spacing.

The initial results indicate that the current output of these irradiated inert gas diodes are not very sensitive to the distance separating the cathode and anode. This surprising result is quite different from those reported for cesium filled thermionic diodes.

Studies on irradiated inert gas diodes as a.c. generators have been made on simple thoriated tungsten filament cylindrical diodes. These tubes contain fission product krypton up to pressures of 100-150 torr. Using their negative resistance properties, it has been shown that these diodes behave like dynatron type oscillators with an upper frequency limit of about 10 megacycles.

2.0. EXPERIMENTS WITH INERT GAS DIODES IN A REACTOR ENVIRONMENT

Six tubes, similar to that described in Figures 1-4 of Quarterly Technical Progress Report No. 1, were constructed and filled with natural argon in the pressure range 27-185 torr. The anode-cathode spacing was 1 mm and the cathode was the standard Philips' impregnated type. These tubes were prepared and tested in the radiation field of the Union Carbide Sterling Forest swimming pool reactor. The purpose of this study was to determine the optimum pressure needed to obtain the maximum output with irradiated thermionic diodes of the above design. The results of these measurements showed that tubes containing 27, 39, 55 and 76 torr of argon were all similar and showed no appreciable difference in current output when operated at a cathode temperature of 1100°C in the core of the nuclear reactor. However, when the pressure was increased to 104 torr and 185 torr, the current output decreased with increasing pressure for the same operating conditions. These data are shown in Figure 1. They compare the different tubes, containing different pressures of argon, at a cathode temperature of 1100°C and a neutron flux level of about 5×10^{13} neutrons/cm^2 sec.

In order to make an equivalent comparison with the above using krypton, five tubes similar to those described above were constructed and filled with natural krypton in the pressure range 10-150 torr. The anode-cathode spacing was 1 mm, the cathode was a Philips' impregnated type cathode and the respective krypton pressures were 150, 70, 40, 22 and 10 torr. The data obtained from four of these tubes when irradiated are illustrated in Figure 2. Although a comparison of the data for 150, 70 and 22 torr would indicate increasing output with decreasing pressure, the curve for 40 torr shows that this is probably not too significant. The data for the tube with a pressure of 10 torr are not included because they are similar to that shown for 70 torr.

It should be noted, however, that the maximum currents obtained with krypton filled tubes (Figure 2) appear to be considerably lower than those for argon filled diodes (Figure 1). The main reason for this result is that the krypton filled tubes were tested in a reactor core hole with a much lower flux density than had been used for the argon filled tubes. The latter were tested in April, 1963 and the former in May, 1963. In the intervening period between these two tests, the Sterling Forest reactor core was changed to increase the burn-up of fuel elements. In this process a fuel element was inserted into the experimental core hole (B-7) which had previously been used. As a result, in the May experiments, another core hole (D-4) was made available. However this new core hole was found to have a lower radiation intensity than the previous one. This is illustrated in Figure 3. Tubes 51B and 53B are identical in that they contain a Philips' type cathode, 100 torr of argon and are constructed like the tube shown in Figure 1 of the Quarterly Technical Progress Report No. 1. It is obvious that the output current in the newer experimental core hole, D-4, is down by at least 40 per cent from that obtained previously. This has to be taken into account if any of the data in Figure 2 are compared with those in Figure 1. This type of comparison indicates that there is no appreciable difference between the use of krypton and argon in tubes of this design.

Another thing which should be noted about the data in Figures 2 and 3 is that negative voltages lead to apparent "negative currents". This very surprising result was observed earlier in 1 mm spaced tubes containing argon. Initially it was felt that this effect occurred because there was leakage across the thin lucalox spacer used in the 1 mm spacing tubes. However, the same result was obtained in an argon filled 1 mm spacing tube when the lucalox spacers were deliberately removed. In addition, reverse currents at negative applied voltages were also found in all the krypton filled diodes of

Figure 2 before they were exposed to radiation. As a result of these measurements it is felt that the following explanation for the phenomenon is the most reasonable. In processing the Philips' cathode, enough barium is emitted from the cathode to make the anode a low work function surface of barium on molybdenum. The proximity of the anode to the cathode (1 mm spacing) ensures that the anode runs quite hot when the cathode is at normal operating temperature. Measurements made on the krypton filled tubes with 1 mm spacing have shown that the anode temperature is about 800°C when the cathode is at 1100°C. Under these circumstances the anode must emit thermionically when a reverse voltage is applied between cathode and anode. This effect of reverse current with applied negative voltage does not occur with diodes whose cathode-anode spacing is 7.5 mm. Such a tube is illustrated by the sketch in Figure 4 and the photograph of Figure 5. In these tubes there is no need to use lucalox spacers to maintain the alignment because of the large cathode-anode spacing. Such a diode was filled with 100 torr of natural krypton and tested in the reactor on the same day as the tubes whose data are shown in Figure 2. The results for this tube at a cathode temperature of 1100°C are shown in Figure 6. In this case, reverse voltages gave zero current. Under these conditions of wide cathode-anode spacing, it is felt that the anode temperature is too low to give any appreciable emission at reverse voltages even though there may be some barium deposited on it. The temperature of the anode of a tube such as illustrated in Figures 4 and 5 has been measured when the cathode was at an operating temperature of 1100°C. These measurements made both in vacuum and at krypton pressures of about 100 torr were found to give a temperature of approximately 400°C.

Since the data of Figure 6 were obtained under similar radiation conditions to those of Figure 2, it is interesting to note that the current from the 7.5 mm spacing tube was higher than that obtained from the 1 mm tube. One possible explanation for the above can be derived from the following argument. Consider the irradiated diode as a cathode and anode separated by a plasma. The current, J, through the plasma is given by

$$J = \rho v_d \qquad (1)$$

where ρ is the ion-electron density in the plasma and v_d is the average drift velocity for an electron traversing the plasma. Increased radiation should increase ρ but not significantly change v_d. Electric field changes in the plasma (e.g., increasing the applied voltage at constant cathode-anode spacing or changing the cathode-anode spacing at a constant applied voltage) would be expected to have a strong influence on v_d but a negligible effect on ρ. However, in the rare gas ambients (Xe, Kr, A), one finds that the current from irradiated inert gas diodes saturates at fairly low voltages (e.g., Figure 6). This can be interpreted to mean that at fairly low field strengths, v_d is relatively constant with increasing electric field, other parameters remaining constant. This phenomenon arises because of the peculiar electron scattering cross section of gases which display the Ramsauer effect. In most cases a decrease of cathode-anode spacing leads to increased electric field strengths for a given anode voltage. In the inert gas diodes, however, v_d shows saturation effects with electric field. This is a possible explanation of why irradiated inert gas thermionic diodes do not show increased output with smaller cathode-anode spacing. Under these conditions the spacing change does not cause any appreciable change in ρ or v_d at moderate voltages.

Equation (1) can be used to offer a possible explanation for increased current output in inert gas diodes with wider spacing. The value of ρ in a particular situation is determined by the rate of ion-electron generation and the rate of ion-electron recombination. The former process is primarily controlled by the intensity of radiation, but the recombination rate is very dependent on geometry. In tubes with large cathode-anode spacing, the rate of recombination should be mainly determined by volume recombination. However, as the separation between cathode and anode becomes smaller, the walls enclosing the volume probably act as recombination centers and tend to decrease the lifetime of the ions and electrons generated by the radiation. Under these circumstances a tube with a smaller cathode-anode spacing would have a lower value of ρ and give less current output than one with a lager cathode-anode spacing.

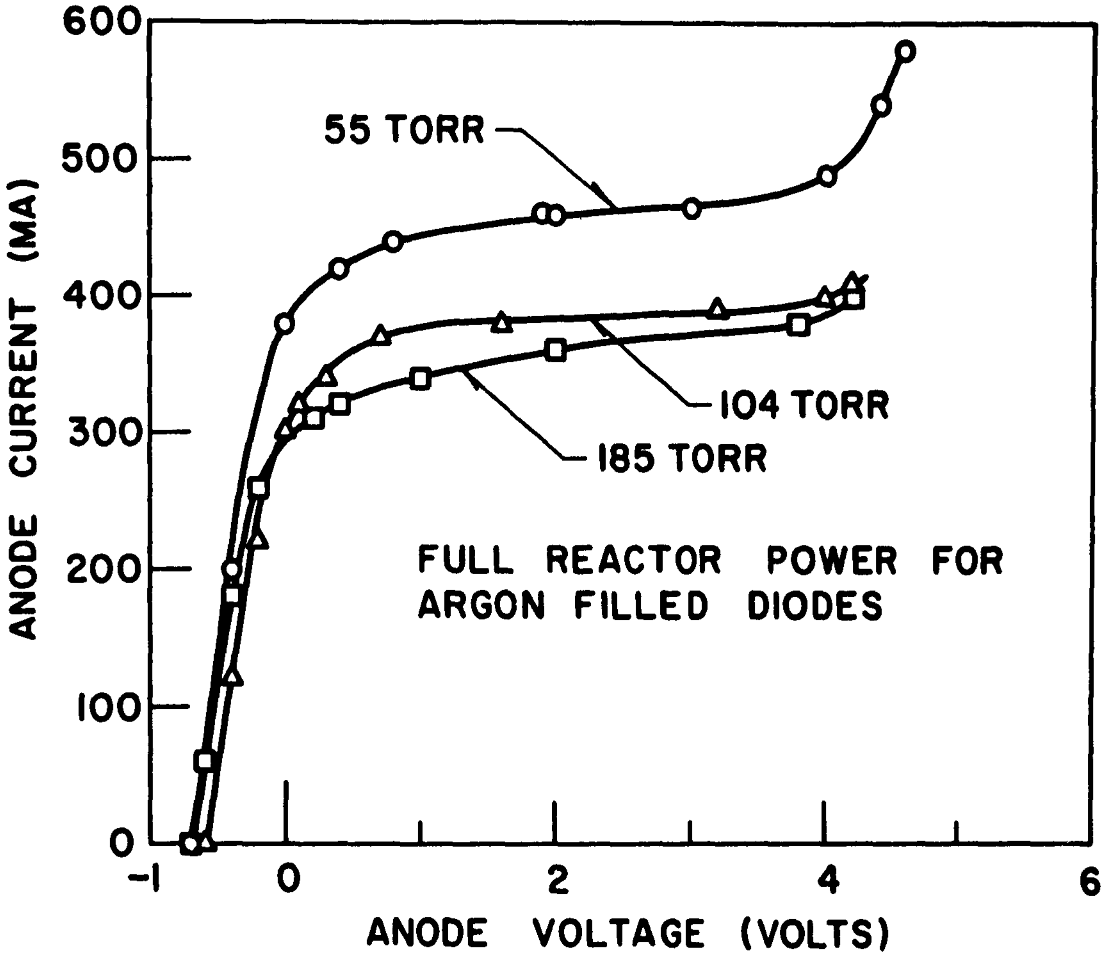
One scheme which has been suggested to increase current output from an irradiated inert gas diode is to increase the radiation dosage above that obtained from the normal gamma radiation in the reactor. To accomplish this a tube has been designed which will lead to increased radiation dosages in the cathode-anode space by the reaction between B^{10} and the neutron flux of the reactor. The (n, α) reaction which results leads to the emission of alpha particles which will further ionize the inert gas between cathode and anode. Such a tube has been constructed and is sketched in Figure 7. A photograph of this tube is shown in Figure 8. The source of B^{10} is the boron nitride spacers between cathode and anode. The natural boron in the boron nitride contains 18.7 per cent B^{10}, which has a very large cross section for thermal neutron capture (~ 4,000 barns). Although boron nitride is an excellent insulator, it was felt that it would be advisable not to have it contact both the anode and cathode in this application. Notice in Figure 8 that there is a space between the cathode and the boron nitride spacer. Tests on these tubes are to be run in the near future.

3.0. STUDIES ON IRRADIATED INERT GAS DIODES AS A.C. GENERATORS

A tube, having planar symmetry, containing an indirectly heated Philips' type cathode and tantalum anode was constructed and filled with fission product krypton to a pressure of about 600 torr. A sketch of the tube, called 44A, is shown in Figure 9. The purpose of this tube was to accomplish a simple proof of principle experiment and show than an a.c. thermionic generator can be made from an irradiated inert gas thermionic diode. The pressure in the tube can be controlled by the cold-finger appendage shown as 7 in Figure 9. The diode displays negative resistance characteristics over a range of pressures from about 30 torr to 150 torr. A typical current-voltage characteristic for this tube at a pressure of 100 torr is shown in Figure 10. Fission product krypton diodes automatically attain low work function anodes after about two weeks. In this interval the decay of Kr^{85} to Rb^{85} causes a monolayer of Rb^{85} to form on the anode surface which is responsible for the formation of a low work function anode. The effect of this process is illustrated in Figure 10 where one can see that there is appreciable current at negative voltages.

The tube 44A was put into the circuit shown in Figure 11. In the external cathode-anode circuit, there is a variable capacitance in parallel with the primary of a 1 : 4 turns ratio transformer (General Radio Type 578-B). Notice that there is no applied voltage in the cathode-anode circuit. The resistor R in the secondary was high impedance in the range of a few megohms. When the gas pressure in space 4 was adjusted to about 100 torr, the circuit in Figure 11 oscillated at a frequency of 400 cycles/sec when C was 1 microfarad and 667 cycles/sec when C was 0.033 microfarads. This experiment shows that a simple a.c. thermionic device is feasible. However it should be emphasized that in this case the power outputs were very low.

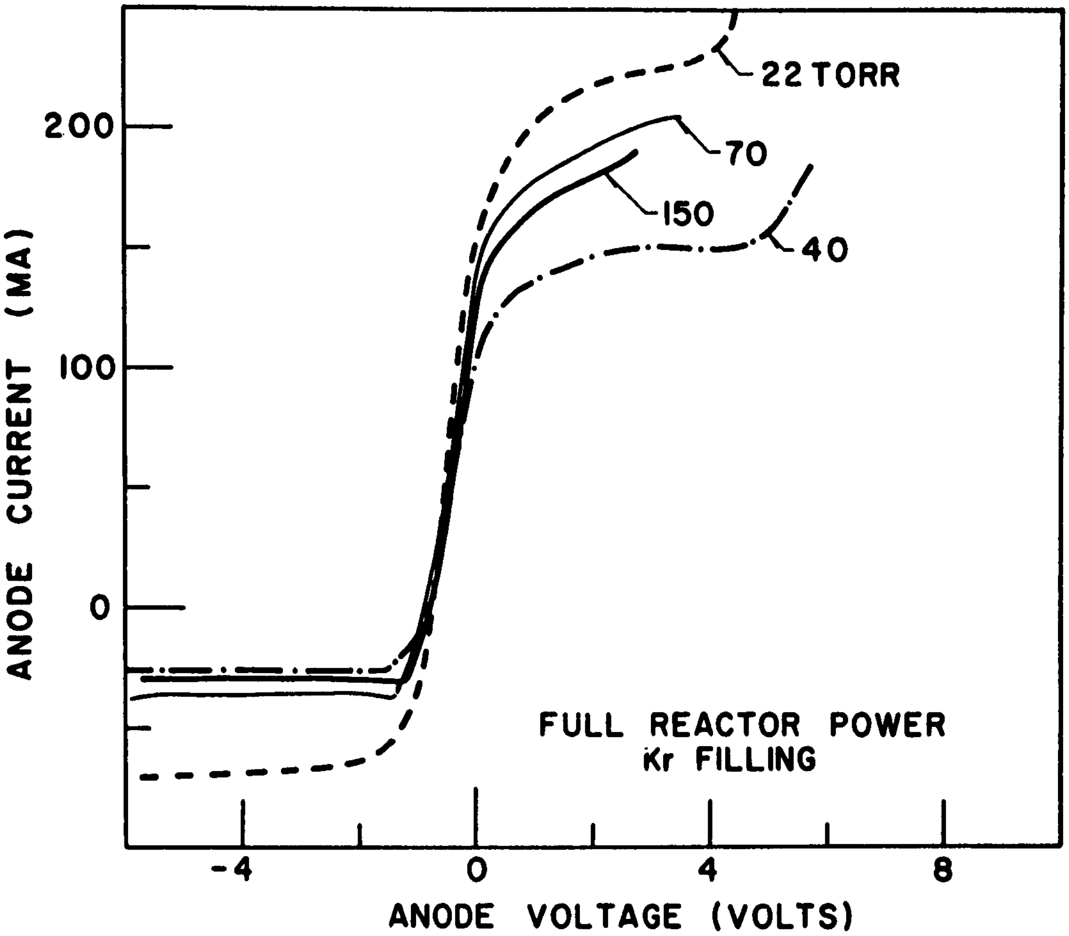
Attempts to measure negative resistance characteristics as a function of frequency are continuing on fission product diodes similar to that shown in Figure 9 of the First Quarterly Report. The purpose of this study is to get a better understanding of electron transport through the inert gas plasma as a function of transit time or frequency. The circuit used in this study is a simple one in which the tube is in series with a 90 ohm resistance and a General Radio high frequency bridge oscillator (Model 1330-A). The voltage across the tube is displayed on the horizontal plates of a Tektronix (Type 536) oscilloscope and the current through the tube, as read by the 90 ohm resistor is measured on the vertical plates. Some difficulties have been experienced in these measurements because of the inherent capacity of the measured diode. At high frequencies the capacity becomes important and this results in the current being out of phase with the voltage. Under these circumstances the retrace lines in the oscilloscope pattern vary considerably from the forward trace as the frequency is increased. Figure 12 illustrates this by showing the oscilloscope patterns as a function of frequency. An attempt is being made to develop a technique for balancing out the capacitance effect. This should permit one to measure the true negative resistance characteristic as a function of frequency.



U-2134

Figure 1. Anode current-voltage characteristics of argon filled irradiated diode placed in the core of a Nuclear Reactor at a neutron flux level of approximately 5 x 10^{13} neutrons/cm^2 sec.

The variable parameter is the pressure of argon in the tube and the cathode temperature is 1100°C.



D-274

Figure 2. Anode current-voltage characteristics of krypton filled irradiated diodes placed in core hole D-4 of the Nuclear Reactor at a neutron flux level of approximately 3 x 10^{13} neutrons/cm^2 sec.

The variable parameter is the pressure of krypton in the tube and the cathode temperature is 1100°C.

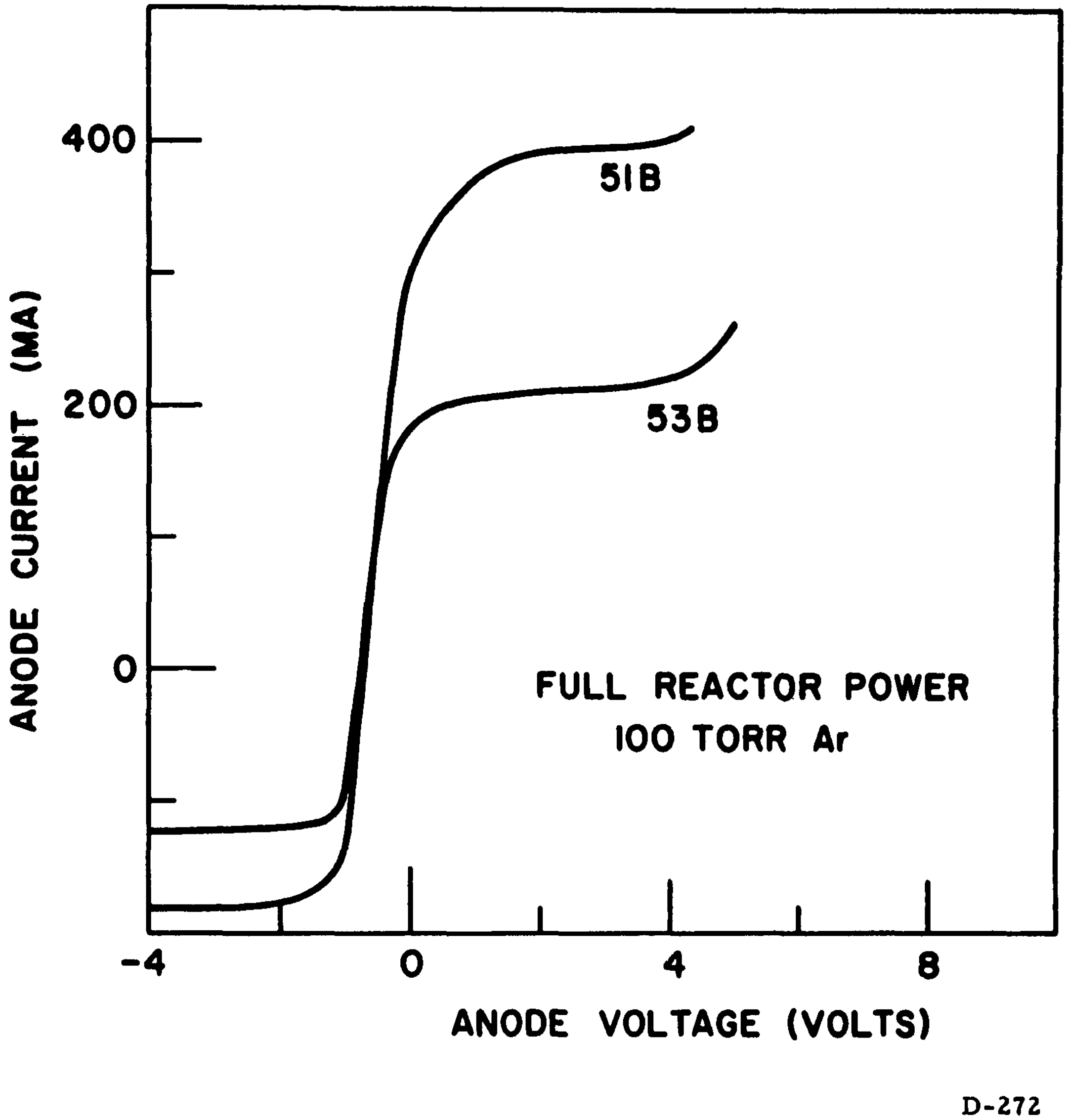
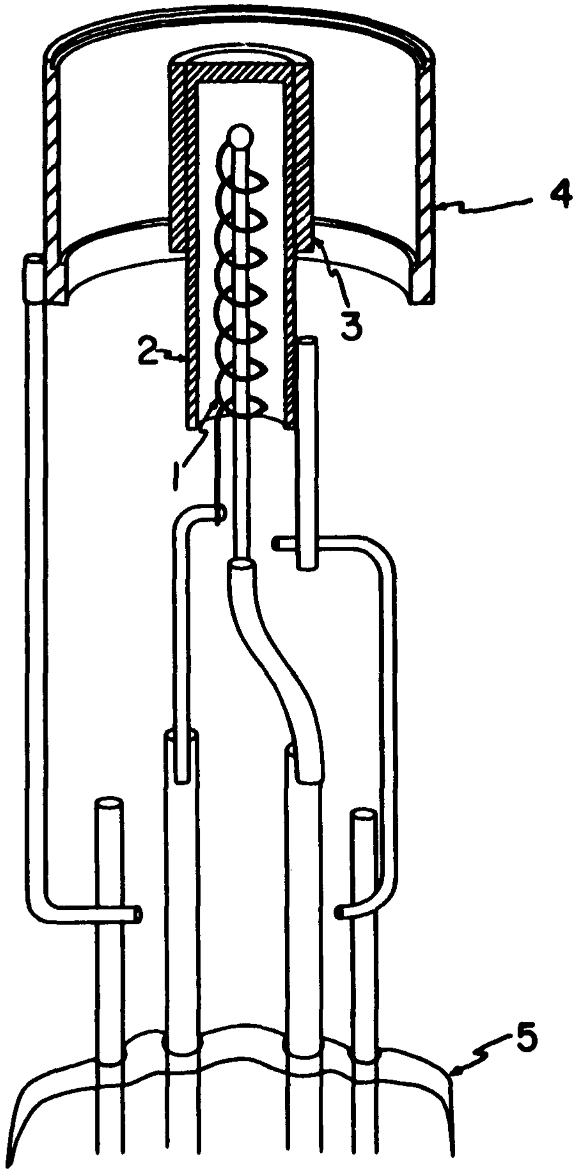


Figure 3. Anode current-voltage characteristics of tubes 51B and 53B.

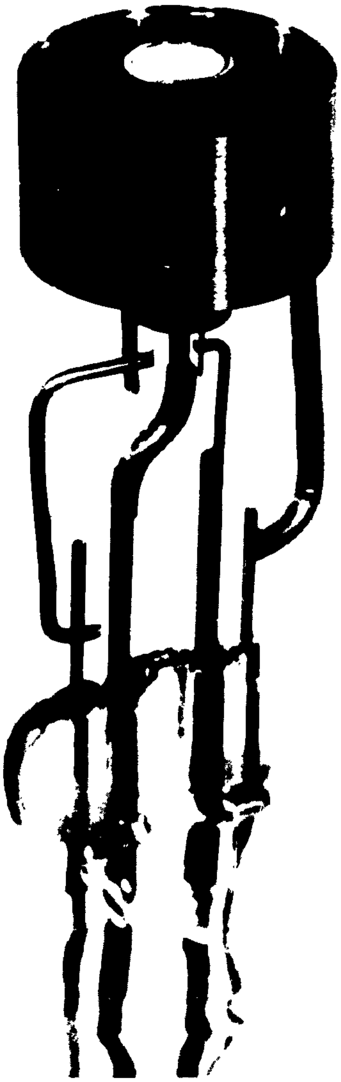
Both tubes were constructed identically. However, tube 51B was tested on 4/20/63 in the old core hole B-7 of the reactor whereas 53B was tested on 5/24/63 in the new experimental core hole D-4. Both measurements were made at the full reactor power of 5 megawatts and at a cathode temperature of 1100°C.



D-299

Figure 4. Sketch of Thermionic Diode with 7.5 mm cathode-anode spacing.

The tungsten filament heater is shown as 1. The molybdenum body of the Philips' cathode is shown as 2. The active impregnated tungsten cylindrical sleeve which partially encases it is shown as 3 and is 0.350 inch in diameter and 0.5 inch long. The molybdenum anode is described as 4 and the glass press as 5. A graded seal from the glass press permits one to seal it to a quartz envelope encasing the tube.



D-302

Figure 5. Photograph of Thermionic Diode with 7.5 mm cathode-anode spacing.

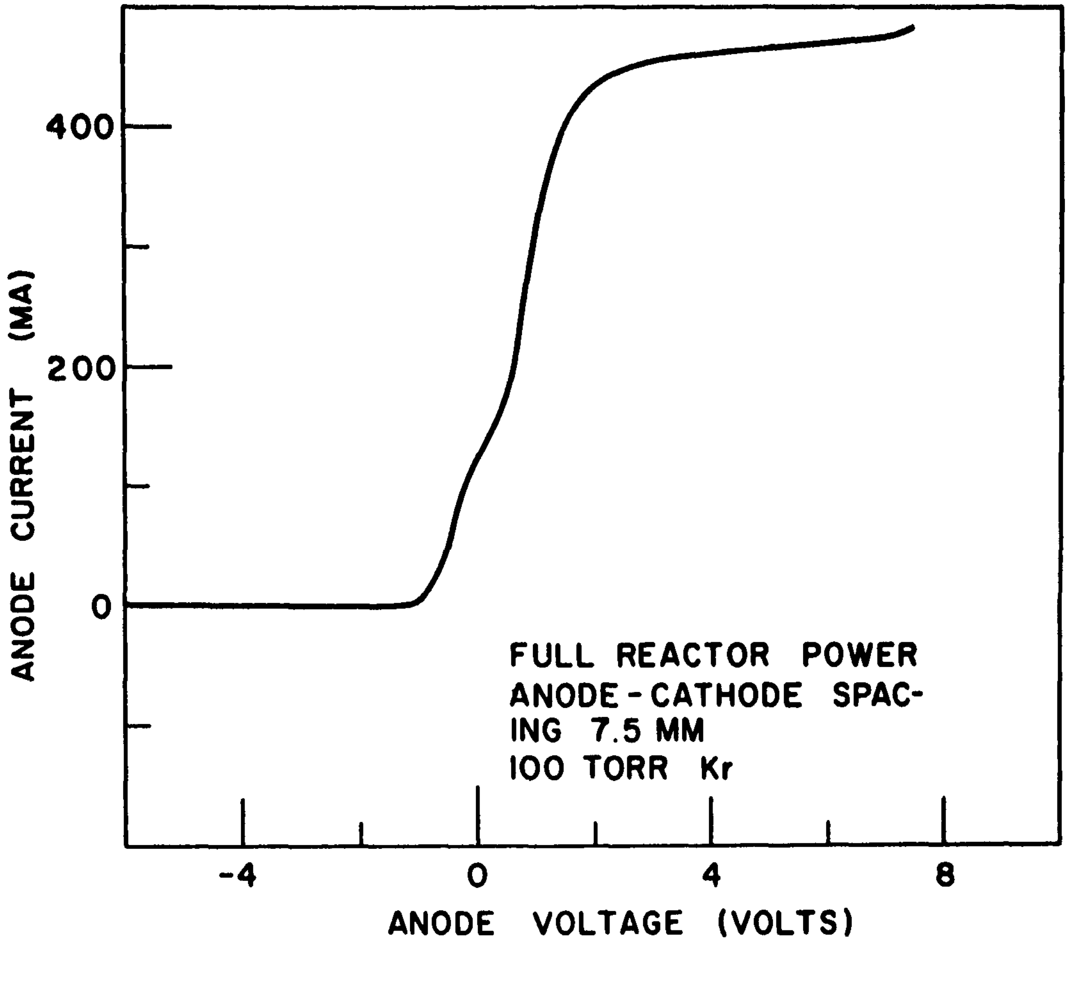
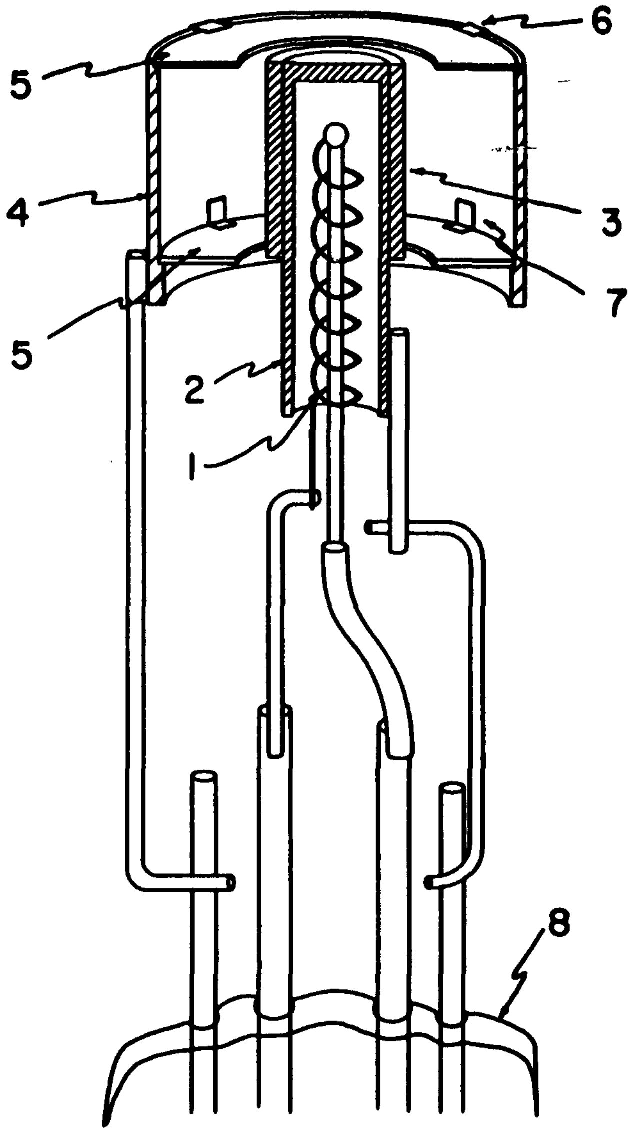


Figure 6. Anode current-voltage characteristic of irradiated diode identical to that shown in Figures 4 and 5 and containing 100 torr of krypton.

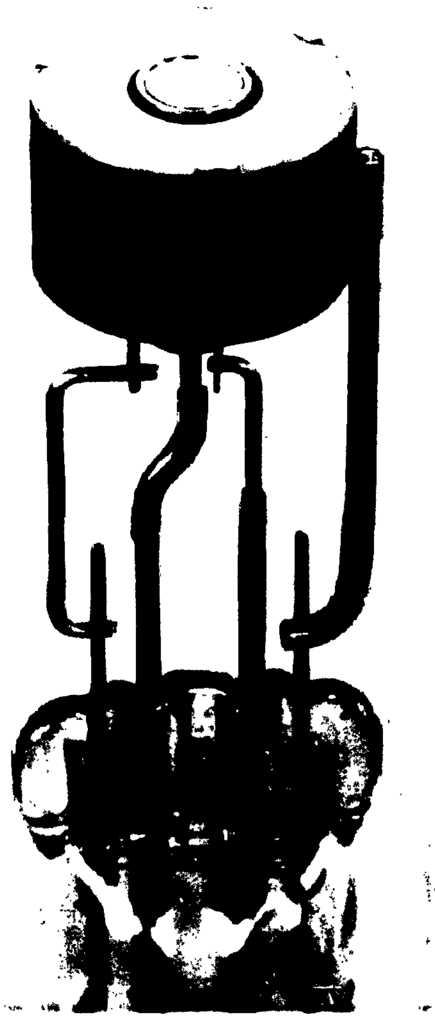
The measurements were made in core hole D-4 of the Nuclear Reactor when it was operating at 5 megawatts of power (neutron flux approximately 3×10^{13} neutrons/cm^2 sec).



D-298

Figure 7. Sketch of Thermionic Diode with boron nitride spacers.

The anode-cathode spacing is 7.5 mm. The tungsten heater filament is shown as 1. The molybdenum body and impregnated porous tungsten cylinder of the Philips' cathode is shown as 2 and 3, respectively. 4 is the molybdenum anode and 5 are the boron nitride spacers which are prevented from moving by the anode ledges they sit on and by tantalum tabs 6 and 7. The glass press through which the supporting leads pass is shown as 8.



D-303

Figure 8. Photograph of Thermionic Diode with boron nitride spacers.

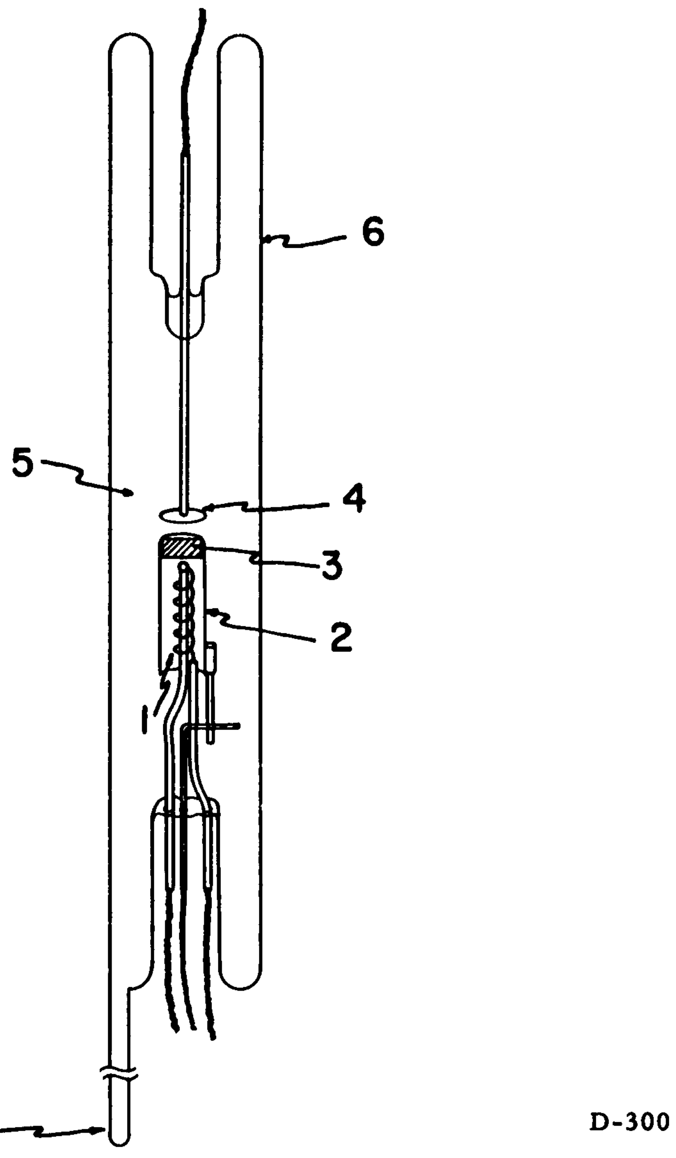
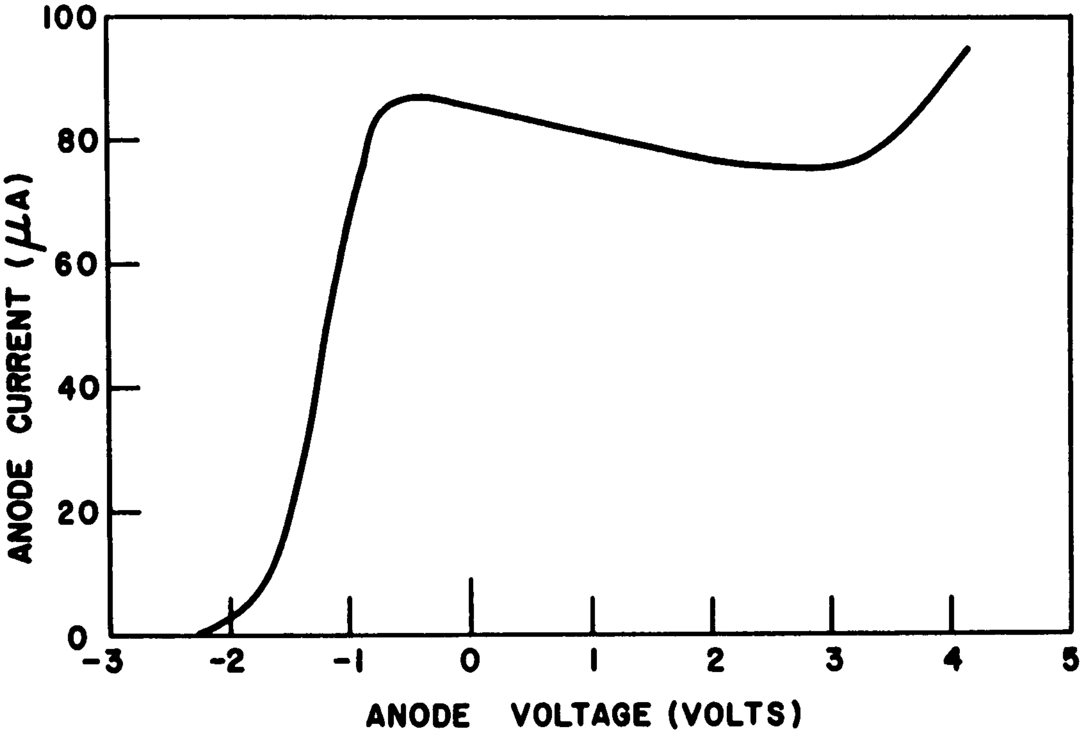


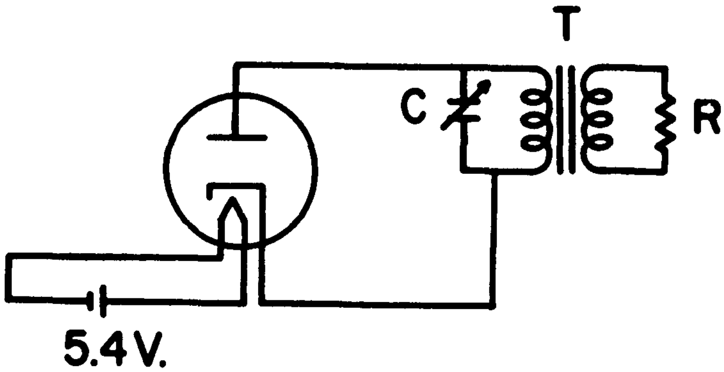
Figure 9. Sketch of planar Thermionic Diode containing fission product krypton.

The anode-cathode space is about 5 mm. The tungsten heater is shown as 1. The molybdenum body of the Philips' type cathode is shown as 2 and the impregnated tungsten porous plug sits on top of the molybdenum body and is shown as 3. 4 is a tantalum disk anode and the space 5 is filled with 600 torr of fission product krypton. 6 is a Pyrex envelope and 7 a cold finger which is used to condense the krypton (using liquid nitrogen) and control its pressure.



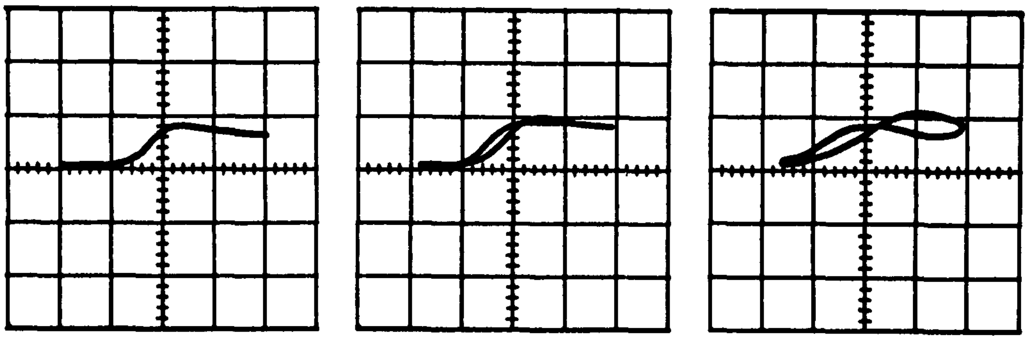
U-2135

Figure 10. Negative resistance characteristics of tube 44A at a fission product krypton pressure of 100 torr.



U-2136

Figure 11. Circuit used to test tube 44A as a.c. Thermionic Generator.



(a) (b) (c)

D-292

Figure 12. Oscilloscope patterns of the negative resistance characteristics of a fission product krypton diode as a function of frequency.

Horizontal scale is 2 volts/division and vertical scale is 5 ma/division. (a) 15 kilocycles (b) 0.15 megacycles (c) 1.5 megacycles.

DISTRIBUTION LIST

Aeronautical Systems Division
Wright-Patterson Air Force Base, Ohio
ASAPR (Library) (1)
ASRPP-20 (5)
ASRNE-4 (1)

Office of Naval Research (1)
Code 429
ATTN: Cmdr. John Connelly
Washington 25, D. C.

AFCRL (CRZAP) (1)
L. G. Hanscom Fld.
Bedford, Massachusetts

SSD (SSTRE, Maj. W. Iller) (1)
AF Unit Post Office
Los Angeles 45, California

Defense Documentation Center (11)
Arlington Hall Stn.
Arlington 12, Virginia

U.S. Army Signal R & D Lab (1)
ATTN: SIGRA/SL-PS (Dr. Kittl)
Ft. Monmouth, New Jersey

U.S. Atomic Energy Commission (1)
Division of Reactor Development
ATTN: Lt. Cmdr. J. Prosser
Washington 25, D.C.

Advanced Research Projects Agency (1)
ATTN: Dr. John Huth
Washington 25, D. C.

Jet Propulsion Laboratory (1)
Spacecraft Secondary Power Section
ATTN: A. Smith
4800 Oak Park Drive
Pasadena, California

Aerospace Corporation (1)
ATTN: Library Technical Document Group
Post Office Box 95085
Los Angeles 45, California

NASA-Lewis Research Center (1)
ATTN: Library, Mr. G. Mandel
21000 Brookpark Road
Cleveland 35, Ohio

NASA Manned Spacecraft Center (1)
SEDD (ATTN: J. D. Murrell)
Houston, Texas

U.S. Atomic Energy Commission (1)
Office of Technical Information Extension
P. O. Box 62
Oak Ridge, Tennessee

Institute for Defense Analysis (1)
ATTN: Dr. R. Hamilton
1825 Connecticut NW
Washington 25, D. C.

Thompson Ramo Wooldridge, Inc. (1)
New Products Research
ATTN: W. J. Leovic
23555 Euclid Avenue
Cleveland 17, Ohio

Thermo Electron Engineering Corporation (1)
ATTN: Dr. G. Hatsopoulos
85 First Avenue
Waltham 54, Massachusetts

General Electric Company (1)
Research Laboratory
Attn: W. Grattidge
P. O. Box 1088
Schenectady, New York

Pratt & Whitney Aircraft (1)
Division of United Aircraft
ATTN: F. A. Corwin
East Hartford, Connecticut

Atomics International (1)
ATTN: Dr. R. C. Allen
P. O. Box 309
Canoga Park, California

Distribution List (Continued)

The Martin Corporation (1)
ATTN: Dr. M. Talaat
Baltimore 3, Maryland

Radio Corporation of America (1)
Electron Tube Division
ATTN: F. G. Block
Lancaster, Pennsylvania

General Motors Corporation (1)
Allison Division
ATTN: D. L. Dresser
Indianapolis 6, Indiana

The Bendix Corporation (1)
Research Laboratory Division
ATTN: George Burton
Southfield, Michigan

Battelle Memorial Institute (1)
R. F. Dickerson
505 King Avenue
Columbus 1, Ohio